Colloid Transport in Porous Media: A Review of Classical Mechanisms and Emerging Topics

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Abstract:

To celebrate the tenth anniversary of Interpore, we present an interdisciplinary review of colloid transport through porous media. This review aims to explore both classical colloid transport and topics that fall outside that purview and thus offer transformative insights into the physics governing transport behaviour. First, we discuss the unique colloid characteristics relative to molecules and larger particles. Then, the classical advection-dispersion-filtration models (both conceptual and mathematical) of colloid transport are introduced as well as anomalous transport behaviours. Next, the forces of interaction between colloids and porous

media surfaces are discussed. Fourth, applications that are interested in maximizing the transport of colloids through porous media is considered. Then the concept of motile, active, biocolloids are introduced and, finally, colloid swarming as a newly-recognized mode of transport is summarized.

1. Introduction

Colloids are nano- or micrometer-sized particles (traditionally defined as between 1nm and 10 μm [1]) which include engineered nanomaterials, viruses, bacteria, fine powders, proteins, and finely textured soil particles (e.g., clay). The transport of colloids through porous media is relevant to a wide range of fields such as medical sciences, industrial manufacturing, geosciences and environmental engineering.

Many industrial processes employ porous media filters to either recover valuable colloidal materials or to purify liquid streams of unwanted materials [1, 2]. Vehicles rely on porous filters to ensure that operation of the fuel, transmission and brake systems are not compromised by colloids [3]. The recovery of valuable metals from waste products, such as catalytic converters found on vehicles or discarded batteries, utilize filtration with porous media as a key step in the recovery process [4-6]. Air filtration in both industrial and consumer products uses porous media to remove airborne colloids [7, 8]. In addition, many systems use emulsions which are an important class of industrial colloids [9].

In medical sciences, colloids such as albumin migrate through the body via porous blood vessels and play a vital role in maintaining human health. While the porous circulatory system is permeable to many molecules, proteins are too large to diffuse across the blood-tissue barrier [10]; this gives albumin the ability to maintain oncotic pressure, i.e. the osmotic pressure that prevents serum from leaking out of capillaries and causing a condition known as edema [11, 12]. In addition, innovative cancer treatment technologies exploit colloid transport behaviour to specifically target cancerous cells [13].

In the geosciences and environmental engineering, there is significant interest in predicting how far colloids such as pathogenic bacteria and viruses travel through soils or other porous media [14-18]. Improper design or maintenance of water treatment filtration systems can result in pathogens entering the water distribution system and cause widespread illness [19]. In

addition, communities utilizing groundwater as a drinking water source rely on the natural soil to act as a filter. There are numerous instances that illustrate how disease outbreaks can occur in communities when pathogens are not adequately filtered during transport through soil [20-24]. Natural geological colloids (e.g., clay particles) are also capable of enhancing the transport of dissolved compounds through porous media as relatively immobile contaminants in the subsurface environment may sorb onto the colloid's surface and be transported further-than-expected distances [25-30].

A large volume of excellent research exists on the transport of colloids through porous media. Field-specific review papers synthesize the current knowledge regarding colloid fate and transport in porous media for medical sciences [10, 31-33], geosciences and environmental engineering [14, 16, 18, 34-40], and industrial manufacturing [2, 3, 41-43]. Other reviews have focused on specific colloid types such as metallic nanoparticles [44-46], or on specific mechanisms such as surface forces [47-51].

The discipline and mechanism-specific reviews currently available in the literature have proven invaluable in synthesizing research and outlining future research needs within their respective areas. However, colloid science researchers would benefit from an interdisciplinary resource that ties together the transport mechanisms that govern colloid transport across a range of scales and disciplines. This review paper aims to fill this gap by providing an introductory resource on the fate and transport of colloids in porous media.

2. Unique Characteristics of Colloids

Colloids have traditionally been defined on the basis of their size. However, differing size ranges appear throughout the literature, depending on the properties of the systems examined. There is no strict definition on what separates a colloid from a molecule (many macro-molecules are considered colloids), or from larger particles. Historically, this delineation has been the subject of some controversy, especially within the biological sciences [e.g., 52]. As a result, it is more useful to define colloids by the properties which influence transport behaviour. Compared to larger particles, colloids exhibit higher rates of diffusive flux (e.g., Brownian motion), higher reactivity due to higher surface areas and are more strongly influenced by short range surface forces. Conversely, colloids possess more deterministic trajectories than smaller molecules, and

their forces of interaction (while based on the same fundamental forces that exist between atom-atom and molecule-molecule pairings) can be larger and decay more slowly across distances than smaller molecules [53]. As diffusivity is inversely proportional to fluid viscosity [54], defining colloids on the basis of transport properties suggests that larger aerosols may possess the same diffusivity of smaller aquasols, but not the same specific surface area or reactivity.

2.1. Brownian Motion

Colloidal diffusion manifests as the movement of colloids from regions of high particle concentrations to regions of low particle concentrations, similar to the molecular diffusion of individual molecules. However, unlike individual molecules, colloids diffuse due to their random walk behaviour, known as 'Brownian motion', which arises due to the large number of collisions that occur between colloids and molecules in the surrounding fluid. The magnitude of Brownian motion increases with decreasing particle size [40].

The Brownian motion of colloids has implications for colloid transport, even in scenarios where the overall diffusive flux is minor relative to the rate of advective transport. Brownian motion alters the trajectories of individual colloids during transport and, ultimately, impacts how far they may be transported through porous media. The trajectory of a single colloid in a flowing fluid can be described using a combination of deterministic (external and hydrodynamic/aerodynamic forces) and stochastic (Brownian force) forces [55, 56].

During transport through a porous medium, a colloid's partially deterministic trajectory causes it to follow flow streamlines through the medium. The Brownian motion aspect of the colloid's trajectory allows it to cross flow streamlines and contact surfaces, or other colloids, that were not on the colloid's initial trajectory [40] (illustrated in Figure S1 of the supplementary information).

2.2. Colloid Surface Area

The specific surface area of particles (i.e. the surface area of a substance per unit mass) increases with decreasing particle diameter. For a single spherical colloid, its surface area (A_s) is given as $A_s = \pi d_p^2$, while its mass (M_s) is $M_s = \frac{1}{6}\pi d_p^3 \rho_s$, where d_p is the colloid's diameter and

114 ρ_s is the colloid's density. The specific surface area (SSA) of a spherical colloid can be described
115 as:

$$SSA = \frac{6}{d_p \rho_s} \quad (2.1)$$

116 This indicates that high SSA is another unique colloid characteristic and may result in colloids
117 being highly reactive. Chemical reactions between colloids and solutes occur on the surface of
118 the colloid [57-59], thus as particle size decreases the total area available for reaction increases,
119 increasing its reactivity [60] and capacity for surface absorption [61]. Reactions may alter the
120 small-scale forces of interaction that govern colloid retention and agglomeration (defined as an
121 assemblage of colloids, not to be confused with pre-nucleation aggregates [62]) .

122 3. Advective-Dispersive Transport and Filtration Behaviour

123 Colloid transport through porous media is classically described at the continuum-scale
124 (i.e., macroscopic-scale) using the classic advection-dispersion partial differential equation
125 (ADE). The ADE typically takes the form described by Eq. 3.1:

$$\frac{\partial C}{\partial t}\phi = -\nabla \cdot (\vec{v}C)\phi + \nabla \cdot \left(\overrightarrow{D_{disp}} \nabla C\right)\phi - \rho_b \frac{\partial S}{\partial t} \quad (3.1)$$

126 where C is the concentration of colloids suspended in fluid ($M \cdot L^{-3}$), t is time (t), ϕ is the
127 medium's porosity, $\vec{v}$ is the pore velocity vector ($L \cdot t^{-1}$), $\overrightarrow{D_{disp}}$ is dispersion tensor ($L^2 \cdot t^{-1}$) , ρ_b is
128 the bulk density of the medium ($M \cdot L^{-3}$)·and S is the mass of colloids retained (i.e. filtered) per
129 mass of porous medium (M colloids·M^{-1} soil). Size exclusion (i.e. colloids unable to enter
130 smaller pores by virtue of their size) may change the effective porosity of the medium and result
131 in colloids travelling at faster-than-expected velocities [63]. The dispersion coefficient represents
132 a non-physical fitting parameter that changes with scale [64]; while this indicates that Eq. 3.1 is
133 not a completely accurate representation of colloid transport, it remains a useful simplification
134 that is utilized for colloid transport through geologic media [40], biological tissues [65], and
135 fibrous filters [3], among others.

136 Generally, colloids are transported in response to gradients of thermodynamic quantities
137 (e.g., colloid diffusion occurs in response to a colloid concentration gradient). As a result, there
138 are numerous other modes of colloid transport from different thermodynamic quantities not

shown in Eq. 3.1 such as: Diffusiophoresis and chemotaxis in which colloids move in response to a solute's gradient [66, 67], electrophoresis [68], magnetophoresis [69], and thermophoresis [70]. In addition, self-propelled colloids are the subject of research for several fields, while many utilize the gradients discussed above [71, 72], motile biocolloids can be mechanically driven via appendages such as flagella [73, 74].

The small-scale interactions between colloids and porous media surfaces govern how far colloids travel via a process known as filtration. The standard conceptual model of filtration is that a porous medium is comprised of pore spaces (i.e., void/fluid phase) and 'collectors' which are discrete units of the solid phase [75]. Colloids are transported through a medium's pore space via advection and dispersion/diffusion until they enter a collector's near-surface zone (typically on the order of nanometers from a collector's surface). Colloids are brought into this near surface zone either when the streamline brings the colloid close to the surface (interception), gravity forces causes the colloid to approach a collector, when Brownian motion carries the colloid across streamlines, or when inertial forces carry the impinging colloid across streamlines and toward the collector [56, 76-80].

The small-scale forces of interaction in the near-surface zone (discussed in section 4) impact whether the colloids are 'retained' by the collector or continue travelling. This process in which colloids are removed from the fluid phase is known as deep bed filtration, which is classically described as a first-order kinetic process with the retention parameter from Eq. 3.1, $\rho_b \frac{\partial s}{\partial t}$, rewritten in terms of:

	$\rho_b \frac{\partial S}{\partial t} = k_{ret} C \phi$	(3.2)

where k_{ret} is the first-order kinetic retention parameter (t^{-1}). Depending on the colloid-colloid forces of interaction, retained colloids may provide further sites for colloid retention, yielding a filter ripening/clogging effect which can decrease medium permeability (i.e., increases a filter's pressure drop) and increase the rate at which colloids are retained [76, 81-84]. For the special case of colloid transport through biological tissues, filtration is a two-step process: 1) adsorption/desorption on the cell surface, and 2) internalization/uptake into the cell body, both of which are describable via first-order processes [85, 86]. Colloid filtration by biological tissue is

166 further limited by the number of available binding /reaction sites on an individual cell [87],
167 conceptually similar to site blocking proposed for geological media [88].

168 3.1. Colloid Filtration Theory

169 There is significant interest in developing mechanistic approaches to estimating k_{ret} (Eq.
170 3.2) to allow a-priori predictions of colloid transport through a porous medium, without relying
171 on empirically determined parameters. The most common approach to mechanistically predicting
172 k_{ret} is colloid filtration theory (CFT), which accounts for the properties of the porous medium,
173 the colloids, the flow regime, and the chemical conditions [56, 75, 76, 80, 82, 89-95]. There are
174 two primary components to modern CFT formulations: 1) a mechanistic model that solves a
175 force/torque balance to estimate the trajectory of a colloid near a collector, and 2) a correlation
176 equation that summarizes the results of the mechanistic model. These results are then upscaled to
177 k_{ret} [40, 96-98]. For a granular medium idealized as a packed bed of Happel Sphere in Cells
178 with diameter d_c, k_{ret} is estimated via Eq. 3.3 [90] where η_o is derived from CFT and represents
179 the fraction of colloids approaching a collector that are retained:

	$k_{ret} = \frac{3}{2}\frac{(1-\phi)}{d_c}\eta_o v$	(3.3)

180 While numerous CFT models exist for granular media in water [56, 80, 90, 91, 99-102], similar
181 models exist for aerosol filtration by fibrous filters which consider flow across a series of
182 cylinder [76, 82, 83, 94].

183 The accuracy of CFT's prediction of k_{ret}, is governed by the small-scale surface forces
184 that dominate colloid-collector interactions in the near surface zone. In scenarios where the
185 colloid-collector surface forces are attractive (i.e., so-called favorable attachment conditions),
186 each colloid-collector contact event results in retention and ADE/CFT generally provides an
187 excellent match to experimental colloid transport data [56]. However, there are scenarios where
188 colloid-collector surface interactions are repulsive and attachment conditions are unfavorable.
189 Unfavourable attachment conditions are predominant in environmental scenarios, as many types
190 of both geologic media and natural colloids carry negative electrostatic charges [40] which yield
191 repulsive forces of interaction. Under unfavourable attachment conditions CFT models are not
192 accurate. The CFT models predict no colloid retention, as the repulsive surface forces prevent

colloid retention [103]. Instead, experimental observations of colloid retention in unfavorable conditions are widespread throughout colloid transport literature [27, 35, 104, 105] which has led to the development of an additional CFT parameter, α, termed attachment efficiency. The attachment efficiency parameter is defined as the fraction of colloids that contact the collector and are retained. Attachment efficiency is incorporated into descriptions of CFT's k_{ret} in Eq. 3.3 as follows [103, 104, 106]:

$$\eta_o = \eta\alpha \quad (3.4)$$

where η is the fraction of colloids approaching the collector that contact the collector (i.e., enter its near-surface zone). Understanding the factors that impact α, and developing predictive models for it, has been the subject of a large number of studies [35, 106-112]. However, no method has successfully demonstrated an ability to predict α a-priori, or fully describe the impacting factors and, as a result, generally remains a fitting parameter.

3.2. Anomalous Transport Behaviour

The first-order retention rate in Eq. 3.2 predicts an exponential retention profile of colloids (i.e., the concentration of retained colloids decreases exponentially with distance). In addition, the ADE predicts a breakthrough curve with a flat plateau and symmetrically sharp increases and decreases in colloid concentration during breakthrough and elution.

Observations of anomalous colloid transport behaviour are widespread. Experimental retention profiles in unfavorable conditions are reported to be hyper-exponential, linearly decreasing or non-monotonic [113-121]. Breakthrough curves may be non-symmetric, a slow increase in concentration over time [122-124], a slow decrease (i.e., filter ripening\clogging) [125], or there may be a slow decrease in colloid concentration during elution (i.e., tailing) (illustrated in Figure S2 in the supplementary information) [126-130]. The application of α to calculate k_{ret} in unfavorable conditions does not resolve the anomaly between experimentally observed anomalous behaviours and the behaviour predicted by the ADE/CFT approach. The predicted retention and breakthrough behaviours are due to the use of k_{ret} as a first-order irreversible retention rate coefficient, applying α as a scaling coefficient for k_{ret} will not change its reaction order or predicted behaviours.

Several theories have been proposed to explain these anomalous behaviours. Non-exponential retention profiles have been explained with theories such as fast and slow colloid retention rates [118], and dual porosities such as mobile and immobile pore space in geologic media [131, 132] or vascular and interstitial pore spaces in biological media [133]. Transport in unfavorable conditions have been hypothesized to be a result of retention at small distances from the collector surface in an energetically favorable region known as the secondary energy minimum [105, 116, 134] or by incorporating zones of discrete micro- and nano-sized zones of favorable attachment conditions on the collector surface [135, 136] and heterogeneous binding sites on cell membranes [137]. In addition, reversible retention and colloid re-entrainment into the bulk pore fluid is considered necessary to produce the observed tailing behaviours [138-140]. However, the macroscopic rate coefficients employed by these models are empirical and can only be determined based on experimental data.

Liquid aerosols also exhibit anomalous transport behaviour. Numerous studies have reported that clogging increases the penetration of liquid aerosols through the filter [141-143], contrary to the decreased penetration observed from solid aerosols. This has been attributed to the aerosol liquid wetting filter fibers [141, 142] as different behaviour is observed under non-wetting conditions [144].

Each CFT model is only valid for a specific porous media and a narrow range of physicochemical conditions [40]. The specific transport mechanisms (i.e., formulation of Eq. 3.1) should be chosen based on desired outcomes and properties of the system; systems with multiple porosities (e.g., biological media) would benefit from a multi-porosity description [133], and weakly retained colloids would require desorption/re-entrainment mechanisms [85], or incorporating transport via gradients of different thermodynamic quantities [71]. In the cases of anomalous transport behaviour, purely mathematical models such as continuous time random walk (CTRW) can accurately match experimental observations [145], but yield little insight into the governing mechanisms. Given the wide variety of models, their varying ranges of applicability and their successes in describing both classic and anomalous colloid transport, the choice of conceptual or computational model to assess colloid transport should not be arbitrary, especially with regards to colloid-collector forces of interaction.

4. Forces of Interaction

As discussed in section 3, the forces of interaction determine how colloids are filtered out of solution and retained by the porous media. As a result, the forces that govern colloid-colloid and colloid-porous media interactions are key to understanding how colloids are transported through porous media and to implementing mechanistic models such as colloid filtration theory.

4.1. Forces of interaction before contact: the DLVO theory

The forces of interaction between colloid-porous media and colloid-colloid interactions are traditionally described using the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory [146].

As colloid-porous media and colloid-colloid interactions are governed by the same forces of interaction, the terms bodies and particles are employed interchangeably throughout this discussion to generically refer to both colloids and porous media. In addition, the term surfaces is used throughout to refer to either colloidal or porous media *surfaces*.

The DLVO model theorizes that the electric charge on surfaces creates an electric potential difference between the surface and the solution, hence attracting oppositely charged ions in solution (counterions). The counterions are simultaneously attracted to the surface (e.g. by electrostatic forces) and dispersed (e.g. thermal Brownian motion). At equilibrium, a counterion concentration gradient exists around the charged surface, with the counterions closest to the surface sensing the strongest attraction to it. The counterion layer around the surface (electrical double layer) can be divided into two layers: 1) the compact Stern layer, mainly subject to the attraction between the counter-ion and the surface, and 2) the shear plane (i.e. the outermost surface), mainly subject to the effect of Brownian motion.

The zeta-potential (i.e. the electric potential at the shear plane) determines the electrostatic interactions between a colloid and other bodies (e.g. porous media). According to the DLVO theory, the zeta-potential (ζ) ($M \cdot L^2 \cdot A^{-1} \cdot t^{-3}$; A denotes current) is related to the charge Q ($A \cdot t$) of the shear plane as follows (Eq. 4.1):

$$\zeta = \frac{4\pi\delta Q}{\varepsilon} \quad (4.1)$$

where δ is the thickness of the diffusive layer (L) and ε is the dielectric constant of the liquid (water).

The double layer (DL) theory has been widely employed to describe the forces of interactions between bodies in fluid. DL forces ($M \cdot L \cdot t^{-2}$) between a sphere and a plane may be for instance expressed as described in Eq. 4.2 [147]:

$$F_{electro} = 128\pi r \gamma_{sphere} \gamma_{plane} n k_b T \kappa^{-1} \exp(-\kappa d) \quad (4.2)$$

where: r = radius of the sphere (L), γ_{sphere} and γ_{plane} are lumped parameters, $\gamma_i = tanh\left(\frac{e\psi_i}{4k_bT}\right)$ where ψ_i is the surface potential (V) and e is the charge of an electron. $n =$ concentration of ions in the bulk solution ($M \cdot L^{-3}$), κ^{-1} = Debye's length (L) and d = distance between the surface of the sphere and the plane (L). The DL force can be attractive or repulsive, depending on the charge of the interacting bodies (e.g. colloids and porous media).

The Debye length measures the range of the double layer. The DLVO theory fails in cell biology applications, when the Debye length is particularly small (< 8 Å; where Å denotes Angstrom), in micellar chemistry applications in which the salt concentrations are high, or in ionic microemulsions or polyelectrolytes systems, for which the screening length is comparable to the hydrated ion sizes [148].

In addition to the DL force, the DLVO theory accounts for van der Waals (VdW) forces and, later, Born forces. VdW forces are always attractive in aqueous media. They represent multipole- multipole interactions and can be classified as follows [149, 150]: 1) Keesom interactions, i.e. the interactions between charges (in the case of molecular ions) and between permanent multipoles; 2) Debye interactions, i.e. the interactions between a permanent multipole with an induced multipole and; 3) London interactions (or dispersion forces), which act between any pair of molecules, regardless of their symmetry.

The VdW interactions are a function of d^{-n} and become infinite when d is zero, whereas DL forces remains finite or rises more slowly than VdW forces [150]. A complete derivation of the expression for VdW forces can be found elsewhere [150]. The VdW forces ($M \cdot L \cdot t^{-2}$) between a sphere and a plane are described by [151]. At short separation distances, the magnitude of VdW forces is always greater than that of DL forces, as described earlier. Therefore, when only VdW and DL forces are considered, the DLVO model predicts infinite attraction at zero separation distance (termed 'primary energy minimum'). Therefore, the DLVO model was expanded to include Born repulsive forces [152] and has been expressed by [151].

The DLVO theory was developed to describe the interactions between hard surfaces, and it is hence not well-suited to describe the interactions between soft charged colloids (i.e., colloids with an ion-permeable surface layer [153]). A new theory was developed by Ohshima to describe the interactions between bodies with a hard core and a soft outer shell [153]. However, the Ohshima model does not account for steric and hydration forces, the importance of which is outlined in section 4.2.

4.2. Non-DLVO forces of interaction before contact: steric, bridging and hydration forces

Steric forces stem from the presence of chains dangling from a surface, e.g. polymer chains attached to colloid surfaces. When polymers overlap, they exert a repulsive osmotic force, the magnitude of which depends on the polymer chemistry and density on the surfaces, on the way they are attached to the surfaces (i.e. if they are physically adsorbed or grafted onto the colloid surface) and on the characteristics of the solvent. In the presence of polymers, bridging forces may also arise because polymers protruding from the surface of a colloidal particle can stick to the surface of a second colloidal particle.

Hydration forces are also not accounted for in the DLVO theory. Hydration forces originate from water structuring at solid-liquid interfaces. In the bulk fluid, hydrogen bonds between water molecules dictate the structure of water. Foreign solutes or colloids introduced in solution affect the hydrogen bonding network because they exclude water molecules, acting as cavities [154]. Small solutes can squeeze between water molecules without breaking any hydrogen bonds. Conversely, colloids break the hydrogen bonds and disrupt the structure of water around the surface of the immersed body [154]. The changes induced in the structure of water depend on the characteristics of the solid surface. In the case of hydrophilic bodies, water molecules become tightly bound onto the surface, preventing the approach of other bodies and producing repulsive interactions [150].

4.3. Relevance of the forces of interaction on colloid transport in geological media

The effect of the forces of interaction on colloid transport in geological media is three-fold: 1) they affect the aggregation of colloids; 2) they determine the rheological behavior of fluids carrying the colloids; 3) they determine if colloids are retained within geological media or migrate in the subsurface. The aggregation behavior of colloidal particles (and particularly of nano-colloidal particles) strongly affects subsurface transport [155, 156]. Attractive forces induce agglomeration, which inhibits transport by clogging the pores, whereas repulsive forces between colloids ensure adequate dispersion and promote their transport. The interplay between hydrodynamic forces and the forces of interaction between colloidal particles and geological media (soil grains or fractured rocks) is also paramount in determining colloid transport [157-164]. Non-DLVO forces play a particularly relevant role when the interacting surfaces are strongly hydrated such as silica and iron [158, 159, 161], when they are soft [159-161, 165], or in the presence of extracellular polymeric substances produced by bacteria [166].

4.4. Forces of interaction and colloid transport in living organisms

Similar to colloid transport in geological media, colloid transport in the mammal and frog body is influenced by the electric charge of the colloids. The importance of electrostatic interactions has been recognized for positive charged colloids interacting with the walls of the gastrointestinal tract [167] and in the context of drug delivery in tumor tissues [168].

Attractive van der Waals forces, steric and depletion forces (e.g. hydrophobic forces), hydration forces (in the case of hydrophilic colloids), and polymer bridging also play a role in the interactions between nanocolloids and circular macromoelcules and biological surfaces [169, 170]. The analysis of the interactions between colloids and biological surfaces in the body is complex because the cells are deformable, their surface is heterogeneous, and they are active [169]. Cells can transport ions and secrete molecules, hence modifying the surface of the colloids they interact with [169]. Nanocolloids in contact with biological fluids become coated with proteins, which can change conformation and lead to either increased affinity between proteins and surfaces or to the exposure of new epitopes (i.e. the part of an antigen molecule to which an antibody attaches itself) [169]. Moreover, specific receptor–ligand interactions can exist and

359 cells can incorporate bound colloids (endocytosis), further increasing the complexity of colloidal
360 transport in the body [169].

361 5. Colloid Injection into Porous Media

362 Many applications of colloid transport in porous media are focused on filtering colloids out
363 of fluids (e.g., potable water filtration), however there is a unique class of applications that are
364 interested in deliberately injecting colloids into porous media. The fate and transport of these
365 injected colloids governs their efficacy, as result there is significant interest in manipulating
366 colloid behaviour in their respective porous environment to maximize transport. These
367 applications include drug delivery into biological tissues [171], enhancing contrast in specific
368 tissues for medical imaging [172], injecting reactive colloids (e.g., nano-zero valent iron; nZVI)
369 [44, 58, 173] or biocolloids [174, 175] into geologic media to remediate soil and groundwater
370 resources, utilizing colloid-sensors for subsurface sensing [176-178] and generating emulsions
371 in-situ for oil recovery [179]. These disparate applications are governed by a similar set of
372 mechanisms (discussed in section's 3 and 4) and significant opportunity exists for cross-
373 disciplinary learning.

374 5.1. Stabilizers and surface coatings

375 Engineered nanoparticles are notoriously unstable (i.e., subject to agglomeration and
376 sedimentation) which can limit mobility [180], inhibit cell uptake in biological tissues [181-184]
377 and reduce colloid reactivity [185]. Colloid flocculation/agglomeration is typically an
378 irreversible process (i.e., kinetically controlled) [186] when colloids interact in the primary
379 energy well; however, in certain conditions colloids may interact in the secondary energy
380 minimum, and agglomeration instead becomes a reversible process [187]. Stabilizers and surface
381 coatings prevent attachment in the primary minimum with repulsive electro-steric forces, in these
382 scenarios reversible attachment governs agglomeration behaviour [185]. As a result, there is a
383 large body of research exploring methods to minimize aggregation and maximize mobility,
384 reactivity and cell uptake in porous media [e.g., 185, 188].

385 Different kinds of stabilizers and surface coatings have been employed to increase the
386 performance (i.e. stability and reactivity) of nZVI [189, 190] and biomedical sensing and nano-

drug delivery [e.g., 191, 192]. For biocolloid applications there is a strong focus on utilizing surfactants to reduce retention by the porous medium [193, 194]. Many of the surfactants studied for reducing bacterial retention [e.g., 195, 196] are similar to those employed in medical and nZVI stabilization, indicating significant potential for cross-disciplinary collaboration.

5.2. Injected Colloid Mobility

Many colloid injection applications report moderate mobility, with transport distances of nZVI and biocolloids ranging from less than 1m to 6m [193, 197-200] and limited penetration of nano-drugs into tumor tissues [201, 202]. Manipulating injection velocities is a promising approach to improving mobility. Retention rates have been observed to decrease with increasing velocity in unfavorable deposition scenarios [203, 204] due to increased hydrodynamic drag [205]. As a result, constant flux methods yield greater travel distances compared to constant head injection [173] due to higher injection velocities. Variations in flow velocity are capable of re-mobilizing retained colloids [206] and potentially improving colloid mobility, yet aside from a pressure pulse technique field trial [207] there is little data available on its efficacy for nZVI injection. This behaviour is not universal to all colloid types, as there is evidence that injecting motile bacteria at high velocities may increase retention [208]. Manipulating velocities is also likely of limited usefulness in scenarios with predominantly favorable attachment conditions, as high flow velocities will be needed to detach colloids from the primary minimum [209]. Manipulating pressure gradients in tumors has had little success in improving colloid transport [e.g., 210].

In addition, mobility can be enhanced using recirculation techniques (e.g., continuously extracting and re-injecting groundwater) or flushing with water or stabilizing solution post-injection [211-213]. Other strategies for improving mobility involve decreasing colloid size via stabilization and synthesis techniques [189], starving biocolloids [214, 215], dynamic size changes [201], or developing biocolloids that either produce biosurfactants or are naturally adhesion resistant [216].

Research focused on chemical locomotion and self-propelled colloids [71, 72, 217] should be considered for aiding colloid mobilization through porous media, either via manipulating gradients, selecting colloids that will self-propel along pre-existing gradients, or designing

colloids with catalytic gradients on their surfaces [e.g., 218]. Additional phenomena such as exclusion zones should be further investigated, as some colloid-surface interactions are capable of generating long range repulsive forces that yield zones up to 100 μm in thickness that are colloid free [219, 220].

6. Particle Swarm Transport in Fractures

Despite decades of colloid transport research, new modes of transport are still being discovered, such as particle swarms which have the potential to transform our understanding of colloid transport through geologic or engineered filters, as well as enable novel approaches to applications that deliberately inject colloids into porous media and seek to maximize their transport. Although the settling speeds of individual colloidal particles is very slow, collective hydrodynamics of colloidal swarms settling under gravity can be much more significant and qualitatively different. Swarms are hypothesized to form in porous media when small, localized clusters of colloids begin to collectively migrate and settle under gravity, such conditions may be found in, among others, slow sand filters and fractured bedrock. In this brief section we explain the origin and scaling of swarm dynamics and indicate how the presence of the surrounding walls of pores or fractures can influence the collective behavior.

A baseline for the behavior of colloidal swarms is provided by the available literature on open-tank conditions [221-228], which is strikingly similar to the behavior of descending vortex rings formed from miscible drops [229-235]. Under conditions of low Reynolds number, colloidal suspensions and miscible drops can be shown to be mathematically equivalent in the gravity-driven Stokes equations [221]: the former represents a kind of approximate discretization of the latter – whether in the laboratory or in a computer simulation. Notwithstanding subtle effects of the granular substructure (fluctuations and modulation of possible influence of fluid inertia [227, 236], a colloidal swarm will therefore behave roughly like a drop of homogeneous miscible liquid with the same density and viscosity as that of the solid-liquid mixture. The analogy of a liquid drop provides the velocity scale: the Stokes settling velocity of an entity of linear dimension d_l and density ρ through an expanse of liquid having density ρ_f and viscosity μ.

$$v_{\text{settling}} = c\frac{(\rho - \rho_f) g {d_l}^2}{\mu}. \quad (6.1)$$

The constant c depends only weakly on the shape and viscosity of the entity. Therefore, it is primarily the characteristic diameter-squared dependence that distinguishes the velocities of individual particles versus swarms. For an individual sphere of diameter d_{p} versus a swarm of diameter d_{s} and (small) volume fraction ϕ, the respective settling velocities are:

$$v_{\text{p}} = \frac{1}{18}\frac{(\rho_p - \rho_f) g d_p^2}{\mu} \propto d_{\text{p}}^2, \quad v_{\text{s}} = \frac{\phi}{15}\frac{(\rho_p - \rho_f) g d_s^2}{\mu} \propto d_{\text{s}}^2 \quad (6.2)$$

Each particle descends with velocity v_{p} (Eq. 6.2) relative to its local liquid environment. Since the liquid environment gets dragged downward by the particles settling nearby, a group of particles will settle faster, v_{s}, than each individual particle. When there are so many particles as to warrant the collective identification as a swarm, then the particles present an additional gravitational force distributed throughout the volume of a grainy pseudo-liquid (i.e., a liquid with an average of the solid- and liquid-phase properties), and the hydrodynamic interactions sum up to produce the characteristic current (a toroidal vortex) inside the swarm, according to the Hadamard-Rybczyinski solution [237-239]. Most important, $v_{\text{s}} >> v_{\text{p}}$ because $(d_{\text{s}}/d_{\text{p}})^2 >> \phi^{-1}$ even for dilute suspensions. For a 1 mm diameter swarm composed of 1% concentration by weight of 1 μm spherical colloids (e.g., clay), there would be ~ 500,000 particles and $v_{\text{s}} \sim 10{,}000\, v_{\text{p}}$. Thus, colloids in a swarm would be transported with speeds much larger than the settling speed of an individual particle.

The complex shape evolution undergone by colloids swarms and vortex rings is well known [221, 222, 224, 227, 230, 231, 240-243] and includes formation of a ring as an intermediate stage. The most interesting and relevant feature is bifurcation of the swarm into two (or more) smaller swarms. This reduction in size immediately slows down sedimentation according to Eq. 6.2. As the presence of even one planar wall [244] reduces the time/distance to breakup, one would anticipate a similar effect for a fracture which is composed of two walls, i.e. rough surfaces. In addition to fractures, swarms have also been observed in granular porous media. A key component of their transport behaviour through both rough fractures and granular media is the swarm's ability to deform in response to changes in aperture. Figure 1 illustrates the

difference in swarm transport through a rough fracture (Figure 1a) in an impermeable matrix compared to a swarm released in a porous medium composed of spherical grains (Figure 1b,c). For rough fractures, bifurcations occur around asperity peaks (Figure 1a). In the cubic packed porous medium, the swarm expands and contracts as it moves from a pore throat into a pore body and back into a pore throat (Figure 1b). The swarm refocuses at every pore body. The repetitive expansion and contraction produced small oscillations in the velocity-depth curve as the swarm asymptotically approaches a constant velocity. Unlike in the fracture, swarm speed can decrease in a porous media from the repetitive reconfiguration of the swarm.

For fractured porous media (Figure 1c), whether or not an intact swarm is transported across a fracture depends on the volume of the swarm, the aperture of the fracture, and the alignment of pores in the matrices across the fracture. Counter intuitively, large-aperture fractures do not enhance swarm transport between porous matrices. Large-aperture fractures result in a decrease in swarm speed, an increase in the probability of swarm bifurcation, and an increase in the probability of particles remaining in the horizontal fracture (Figure 1c). Large-aperture fractures caused this reduction in speed because a swarm is free to expand laterally in the fracture, increasing the swarm diameter and the drag on the swarm.

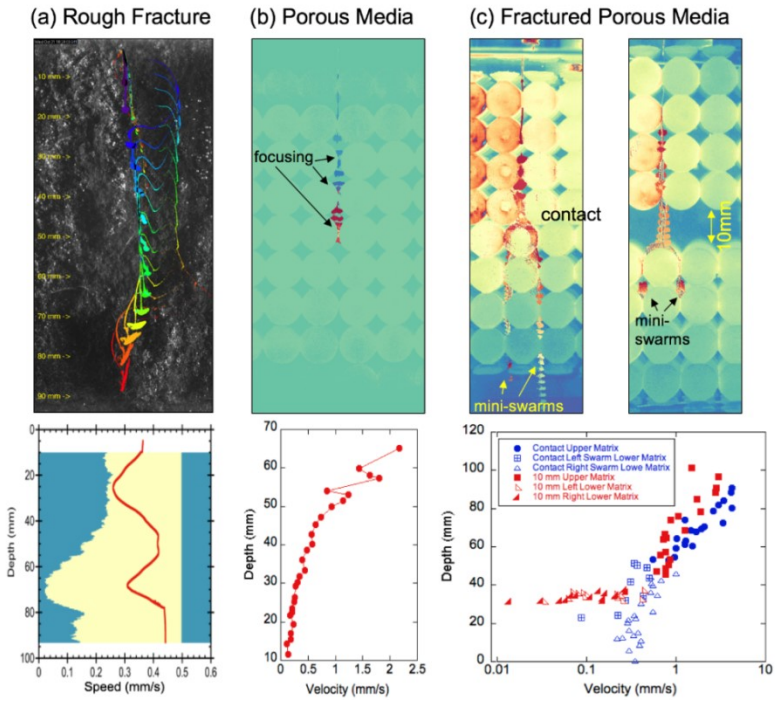

Fig. 1 (top row) Time-lapsed composite images and (bottom row) swarm velocity as a function of depth for a swarm in (a) a rough-walled fracture, (b) a cubic packed porous medium, and (c) fractured porous media with the fracture in contact and for a 10 mm aperture, showing swarm deformation in response to changes in confinement geometry.

7. Active Microbiological Colloids

Much of the discussion above has focused on the transport of passive colloids (i.e., colloids incapable of self-propulsion). However, in nature numerous colloids are capable of self-propelled transport (known as active colloids [245-248]) such as sperm inside the female reproductive tract where thousands of cells move through narrow channels in harsh acidic environments [249, 250]. Other examples include the decontamination of waste water through biofilters or slow sand filters. In these systems, biofilms can form at the top of the filter, allowing the retention of both particles and self-propelled microorganisms [251-255]. Active colloids also exist in the field of medicine; the implantation of biocompatible scaffolds allows cells, such as osteoblasts, to colonize porous material and promote tissue regrowth [256]. Even gut microbiota can be considered as a complex system able to contribute to nutrient absorption or immune system responses [257, 258]. These active colloids are not in thermodynamic equilibrium and their study can reveal new mechanisms linking biological processes with physical laws.

7.1. Interactions between swimmers and surfaces or interfaces

Active swimmers may be classified according to the force that they exert on the fluid. Considering far-field hydrodynamics, the simplest model is represented by a dipole force [259-261], and depending on whether the dipole is extensile or contractile, this leads to two categories: pushers and pullers, respectively (illustrated in Figure S3 of the supplementary information). From this point of view, Escherichia coli or Bacillus subtilis are pushers, and algae such as *Chlamydomonas reinhardii*, can be considered pullers. These two categories do not fully cover all the possibilities of propulsion at low Reynolds numbers [247, 262-265], but help distinguish two types of effects. Organisms such as *Paramecium* with complex cilia mechanics cannot be described as a dipole [266, 267].

513 *Escherichia coli* [268] represents a good model for active colloids[269]. Its body has an
514 ellipsoidal-shape (1 μm diameter and 2 μm length), where the motion is the consequence of the
515 rotation of several helicoidal flagella. Each flagellum is linked to the cell membrane by a
516 nanoscale motor. When all the motors turn counter-clockwise, the flagella rotate in a bundle and
517 push the cell forward. When a motor rotation switches to clockwise, the cell can change
518 direction. When a pusher-like swimmer approaches a solid, it will remain swimming parallel to
519 the wall, producing a long dwelling time or a trapping mechanism at the surface. This
520 phenomenon will generate an increase of bacterial density in vicinities of a solid [270-274]. The
521 flagella or cilia structure can contribute to scattering from the wall [275], despite other evidence
522 that scattering is caused by hydrodynamics [276]. In the absence of flow, interaction with
523 spheres and cylindrical objects is also observed [277, 278]. Puller-type swimmers scatter from
524 curved surfaces, and both hydrodynamic and steric forces are necessary to explain such
525 interactions [279].

526 7.2. Active Bacteria-Fluid Interactions

527 It is well known that fluid properties can modify the motility of microorganisms.
528 However, later work has also demonstrated that biologically active particles can modify fluid
529 properties. Living particles in a suspension create a non-equilibrium system where organisms are
530 constantly modifying their surroundings by taking in nutrients and secreting waste [280] such as
531 with fermentation processes, where the production of alcohol reduces fluid density and alters pH.
532 Self-propelled cells can modify fluid rheological properties [281, 282] and exhibit non-
533 Newtonian behavior [283]. Pusher-type swimmers can decrease viscosity [283, 284], flagellar
534 beating as in puller-type swimmers can increase viscosity [285, 286]. Under certain conditions,
535 active colloids can even achieve the characteristic of frictionless superfluids [287, 288].

536 In addition, flowing fluid can modify bacterial transport behavior [289]. In small
537 channels, high shear forces near walls can re-orient bacteria and result in upstream movement
538 [290, 291]. In addition, active bacteria tend to move from channel centers to the high shear
539 region near lateral solid surfaces [292, 293]. Flow also influences bacterial quorum sensing (i.e.
540 chemical communication) that leads to collective responses and biofilm formation [294].

541 7.3. Microbiological Transport in Porous Media

In porous media, the flow and shear rate distribution will yield a complex combination of the behaviours discussed above. Bacteria are capable of moving through pores sizes smaller than their body size [295]. Magnetotactic bacteria are able to travel effectively through heterogeneous porous media and overcome relatively high pore water velocities (~250μm/s) via magnetotaxis [296]. Bacteria also accumulate on the downstream stagnation points of collectors where flow velocities are smaller than bacteria speed [297]. Despite this work, the influence of motility on retention in porous media remains poorly understood [298]. The complexity of interactions between self-propelled microorganisms and porous media provides substantial opportunities for future research and a valuable avenue for understanding transport processes in porous media and optimizing applications that aim to either limit or maximize colloid mobility in porous media.

8. Conclusions

Despite decades of research into colloid transport through porous media, substantial future research opportunities exist. New mechanisms, phenomena and modes of transport are still continuously being revealed. The origins of many anomalous transport behaviours and colloid-colloid and colloid-collector interactions are still unknown. In addition, new colloid types such as engineered self-propelled colloids and applications such as biomedical and subsurface sensing ensures that colloid transport through porous media will remain a rich and diverse field for many decades yet.

While colloid transport research is conducted across a wide range of disciplines and porous media types, this paper has outlined how the physics governing colloid transport and approaches to solving colloid-related problems, are remarkably similar. For instance, the efficacy of engineered nanoparticles for use in medicine and geosciences are limited by their agglomeration and poor mobility. Similar approaches are being undertaken to improve their delivery, such as manipulating injection conditions and stabilizing particles utilizing electro-steric forces. As a result, cross-discipline collaborations and research will prove invaluable to solving the myriad of outstanding problems in colloid transport through porous media.

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